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NASA TECH BRIEF

Lewis Research Center

Single Crystal Tubes of Beta Alumina

Single crystal tubes of sodium beta alumina have been produced for the first time. Prior art did not permit the growth of either tubular shapes or large single crystals of beta-alumina. However, a process called edge-defined, film-fed growth allows both tubular shapes and single crystallinity to be achieved.

Sodium beta alumina is a solid state electrolyte membrane material suitable for use in high energy density batteries such as the sodium sulfur battery. The availability of beta alumina in single crystal form as opposed to plates of pressed and sintered powder would make possible membranes with improved conductivities. Such single crystal membranes would also eliminate problems associated with the electrical short circuiting of the membrane due to possible sodium metal diffusion along grain boundaries or through interconnected pores. One favorable geometry for such single crystals is tubular, with the high ionic conductivity direction of the membrane normal to the tube axis. Such properly oriented tubular single crystals of beta alumina may permit the construction of batteries with a longer lifetime, lower operating temperature and/or higher energy density.

There are, however, considerable difficulties associated with the crystal growth of sodium beta alumina. These difficulties are due principally to the fact that beta alumina appears to decompose peritectically (i.e., change composition in the solid-liquid-solid cycle), as well as to the high vapor pressure of sodium over beta alumina at its decomposition temperature. Single crystal tubes (5 mm (0.2 inch) O.D., 3 mm (0.12 inch) I.D., 25-35 mm (1-1.4 inches) long) of sodium beta alumina have finally been fabricated by a process combining edge-defined, film-fed growth (EFG) and the use of excess Na_2O as a flux, together with up to 300 psi of an inert gas overpressure, to achieve the desired single crystallinity and chemical composition.

Edge-defined, film-fed growth is a process by which a single crystal or dense polycrystal body can be formed having a shape controlled by the outside dimensions of a die, the growth actually taking place from an extremely thin film of liquid fed by capillary action from a crucible below. Since beta alumina appears to decompose peritectically, it is necessary to grow this material from a melt containing an excess of Na_2O in order to keep the growth temperature less than the peritectic decomposition temperature. Furthermore, because of the high vapor pressure of sodium over such melts, it is necessary to use a high inert gas overpressure in order to suppress sodium loss during the crystal growth process.

For the growth of tubular sodium beta alumina crystals, the following procedure is used: the crucible, EFG tube die, and charge are placed in a pressurized chamber capable of applying an overpressure of 300 psi of argon. When the crucible and charge are heated to produce melting, the liquid melt rises by capillary action to fill the feeding orifice in the die. A small heat sink or seed is then brought into contact with the melt at the top of this capillary feed slot. After adjustment of the melt temperature and seed withdrawal rate, the melt spreads across the top surface of the die, until further spreading is prevented by the 90° change in contact angle at the edges of the die. The growth of a tubular shape from a thin liquid meniscus is then established. The use of melt compositions enriched in Na_2O enables this growth process to be carried out at temperatures below the decomposition temperature of sodium beta alumina.

Notes:

1. The edge-defined, film-fed growth process was developed by Tyco Laboratories, Inc., 16 Hickory Drive, Waltham, Massachusetts 02154.
2. Further information is available in the following report:

   NASA CR-121033 (N73-12810), Development of Single Crystal Membranes

   Copies may be obtained at cost from:

   Aerospace Research Applications Center
   Indiana University
   400 East Seventh Street
   Bloomington, Indiana 47401
   Telephone: 812-337-7833
   Reference: B73-10316

(continued overleaf)

3. Specific technical questions may be directed to:
Technology Utilization Officer
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Patent Status:
NASA has decided not to apply for a patent.

Source: R.W. Stormont, F.H. Cocks,
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Tyco Laboratories, Inc.
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